

Power functional theory for Brownian dynamics

Matthias Schmidt¹ and Joseph M. Brader²

¹*Theoretische Physik II, Physikalisches Institut, Universität Bayreuth, D-95440 Bayreuth, Germany*

²*Department of Physics, University of Fribourg, CH-1700 Fribourg, Switzerland*

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For classical many-body systems subject to over-damped Langevin dynamics, we propose a dynamical functional consisting of reversible free energy changes and irreversible power dissipation. Minimization with respect to the microscopic one-body current yields a closed equation of motion. Dynamical density functional theory is obtained when using the ideal gas power functional. Approximations to the excess (over ideal) power functional yield numerically tractable equations of motion beyond the adiabatic approximation, opening the door to the systematic study of systems far from equilibrium.

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The dynamics of soft matter is a topic of considerable current interest, both from an experimental and theoretical perspective [1, 2]. Among the various theories aiming to provide a microscopic description of complex transport and pattern formation phenomena, the dynamical density functional theory (DDFT) [3, 4] has emerged as a prime candidate. Much of the appeal of DDFT arises from its close connection to equilibrium density functional theory (DFT), which is a virtually unchallenged framework for the study of equilibrium properties in inhomogeneous liquids [3].

The DDFT was originally suggested on the basis of phenomenological reasoning [3], and has since been red-erived from the Langevin equation [4], using projection operators [5] and via coarse graining the many-body Smoluchowski equation [6]. Despite the deep insight provided by these alternative derivations of the DDFT, the final equation of motion for the density remains unchanged and no guidance is provided for making a systematic improvement that goes beyond the standard formulation. A striking and undesirable feature of DDFT is the clear asymmetry between the treatment of spatial and temporal degrees of freedom. The intricate nonlocal spatial structure is in stark contrast to the simple time-local treatment of the dynamics and suggests much potential for improvement, particularly in view of the sophistication of memory function approaches such as mode-coupling theory [7].

At the heart of DDFT lies the ‘adiabatic approximation’, in which the nonequilibrium pair correlations of the real system are approximated by those of a fictitious equilibrium, whose density distribution is given by the instantaneous density of the nonequilibrium system [8]. Although reasonable for describing relaxational dynamics close to equilibrium, this assumption fails when applied to even the simplest driven systems (e.g. shear flow [9]), thus putting out of reach many relevant and fundamental nonequilibrium problems.

In this Letter, we overcome the above limitations by developing a variational theory based on the current

rather than the density as the fundamental variable. Our framework transcends the adiabatic approximation and provides a physically intuitive method to go beyond DDFT.

The central variable in DDFT is the time-dependent one-body density,

$$\rho(\mathbf{r}, t) = \left\langle \sum_i \delta(\mathbf{r} - \mathbf{r}_i(t)) \right\rangle, \quad (1)$$

where $\langle \cdot \rangle$ indicates an average over trajectories, $\mathbf{r}_i(t)$ is the position of particle i at time t , $\delta(\cdot)$ is the Dirac distribution, and the sum is over all N particles in the system. However, the motion of the system is better characterized by the time- and space-resolved one-body current

$$\mathbf{J}(\mathbf{r}, t) = \left\langle \sum_i \mathbf{v}_i(t) \delta(\mathbf{r} - \mathbf{r}_i(t)) \right\rangle, \quad (2)$$

where $\mathbf{v}_i(t) \equiv d\mathbf{r}_i(t)/dt$ is the velocity of particle i . It is evident that the vector current (2) provides a more appropriate starting point than the density (1) for a theory aiming to describe flow in a complex liquid; there are many relevant flows for which the density is trivial, $\rho(\mathbf{r}, t) = \text{const}$, but the current is not. Once the current is known, the density can be calculated from the continuity equation,

$$\rho(\mathbf{r}, t) = \rho(\mathbf{r}, t_0) - \int_{t_0}^t dt' \nabla \cdot \mathbf{J}(\mathbf{r}, t'), \quad (3)$$

which expresses the local conservation of the particle number. Prior to the initial time t_0 we take the system to have been in equilibrium.

Starting from a particle-based description, we write the (over-damped) Langevin equation as

$$\gamma \mathbf{v}_i - \mathbf{F}_i = 0, \quad (4)$$

where γ is a friction constant. The viscous force is balanced by the total force

$$\mathbf{F}_i = -\nabla_i U(\mathbf{r}^N) - \nabla_i V_{\text{ext}}(\mathbf{r}_i, t) + \mathbf{X}(\mathbf{r}_i, t) + \boldsymbol{\chi}_i(t), \quad (5)$$

consisting of four contributions: (i) Forces arising from the total interaction potential, $U(\mathbf{r}^N)$. (ii) Conservative forces generated by an external potential, $V_{\text{ext}}(\mathbf{r}, t)$. (iii) Non-conservative forces $\mathbf{X}(\mathbf{r}, t)$. (iv) Stochastic, thermal forces, $\chi_i(t)$, which are auto-correlated according to $\langle \chi_i(t) \chi_j(t') \rangle = 2k_B T \gamma \delta_{ij} \delta(t - t') \mathbf{1}$, where k_B is the Boltzmann constant and T is temperature.

In the spirit of the Rayleigh dissipation function [10], but including both dissipative and conservative forces, we define a generating function

$$R(\mathbf{r}^N, \mathbf{v}^N) = \sum_i \left(\frac{\gamma}{2} \mathbf{v}_i - \mathbf{F}_i \right) \cdot \mathbf{v}_i, \quad (6)$$

obtained by integrating the left hand side of (4) formally with respect to \mathbf{v}_i and summing over all particles. Clearly, the Langevin equation (4) is then given by

$$\frac{\partial R(\mathbf{r}^N, \mathbf{v}^N)}{\partial \mathbf{v}_i} = 0, \quad (7)$$

where the derivative is taken at fixed particle positions, $\mathbf{r}^N = \text{const}$; the physically realized velocities \mathbf{v}^N minimize R . When averaged and integrated over time the function R plays the role of a generating *functional* for the density (1) and current (2) via

$$\rho(\mathbf{r}, t) = -\frac{\delta}{\delta \dot{V}_{\text{ext}}(\mathbf{r}, t)} \int dt \langle R \rangle, \quad (8)$$

$$\mathbf{J}(\mathbf{r}, t) = -\frac{\delta}{\delta \mathbf{X}(\mathbf{r}, t)} \int dt \langle R \rangle, \quad (9)$$

where $\dot{V}_{\text{ext}}(\mathbf{r}, t) = \partial V_{\text{ext}}(\mathbf{r}, t) / \partial t$. We can hence obtain the intrinsic part of the generator $\int dt \langle R \rangle$ as a functional of $\rho(\mathbf{r}, t)$ and $\mathbf{J}(\mathbf{r}, t)$ via Legendre transform

$$W[\rho, \mathbf{J}] = \int dt \langle R \rangle + \int dt \int d\mathbf{r} \left(\mathbf{X}(\mathbf{r}, t) \cdot \mathbf{J}(\mathbf{r}, t) + \dot{V}_{\text{ext}}(\mathbf{r}, t) \rho(\mathbf{r}, t) \right), \quad (10)$$

which we split into a sum of dissipative and reversible contributions,

$$W[\rho, \mathbf{J}] = \int dt \left(P[\rho, \mathbf{J}] + \dot{F}[\rho] \right). \quad (11)$$

Here the integrand consists of a dissipated power functional, $P[\rho, \mathbf{J}]$, accounting for irreversible energy loss due to the friction, and the total time derivative (indicated by the dot) of the intrinsic Helmholtz free energy functional, $F[\rho]$, responsible for reversible free energy changes [3]. In general, the dissipation functional $P[\rho, \mathbf{J}]$ will be non-local in space and time, depending on the history of the fields $\rho(\mathbf{r}, t)$ and $\mathbf{J}(\mathbf{r}, t)$ prior to time t . Employing the functional chain rule, $\dot{F}[\rho] = \int d\mathbf{r} \dot{\rho}(\mathbf{r}, t) \delta F[\rho] / \delta \rho(\mathbf{r}, t)$, and the continuity equation (3), followed by a partial integration in space leads to

$$W[\rho, \mathbf{J}] = \int dt \left(P[\rho, \mathbf{J}] + \int d\mathbf{r} \mathbf{J}(\mathbf{r}, t) \cdot \nabla \frac{\delta F[\rho]}{\delta \rho(\mathbf{r}, t)} \right). \quad (12)$$

This intrinsic contribution can be combined with the external work in order to define a total dissipation functional

$$\mathcal{R}[\rho, \mathbf{J}] = W[\rho, \mathbf{J}] - \int dt \int d\mathbf{r} (\mathbf{X}(\mathbf{r}, t) - \nabla V_{\text{ext}}(\mathbf{r}, t)) \cdot \mathbf{J}(\mathbf{r}, t). \quad (13)$$

Recognizing (7), an equation for the time evolution of the system is obtained [11] by

$$\frac{\delta \mathcal{R}[\rho, \mathbf{J}]}{\delta \mathbf{J}(\mathbf{r}, t)} = 0, \quad (14)$$

at $\rho(\mathbf{r}, t) = \text{const}$. The result is our fundamental equation of motion

$$\int dt' \frac{\delta P[\rho', \mathbf{J}']}{\delta \mathbf{J}(\mathbf{r}, t)} = -\nabla \frac{\delta F[\rho]}{\delta \rho(\mathbf{r}, t)} - \nabla V_{\text{ext}}(\mathbf{r}, t) + \mathbf{X}(\mathbf{r}, t), \quad (15)$$

where the temporal variable in (11) has been relabelled t' , and we used the shorthand notation $\rho' = \rho(\mathbf{r}, t')$ and $\mathbf{J}' = \mathbf{J}(\mathbf{r}, t')$; furthermore $\delta P[\rho', \mathbf{J}'] / \delta \mathbf{J}(\mathbf{r}, t) = 0$ for $t > t'$ due to the fact that $P[\rho', \mathbf{J}']$ depends only on the value of its arguments at times prior to t' , cf. the discussion of this causal structure below (11).

The term on the left hand side of (15) represents a friction force, balanced by the terms on the right hand side arising from inhomogeneities in the local intrinsic chemical potential, $\delta F[\rho] / \delta \rho(\mathbf{r}, t)$, and external forces. In equilibrium the left hand side of (15) vanishes and the variational prescription reduces to $\delta \Omega[\rho] / \delta \rho(\mathbf{r}) = 0$, where $\Omega[\rho]$ is the grand potential functional [3], thus recovering equilibrium DFT as a special case. In nonequilibrium, the challenge is to find explicit forms for the dissipation functional, $P[\rho, \mathbf{J}]$, to obtain a closed equation of motion. To gain some intuition we consider three special cases:

(i) *Frictionless motion.* The simplest case is obtained by neglecting nonconservative external forces and setting $P[\rho, \mathbf{J}] = 0$. Equation (15) then yields $\delta F[\rho] / \delta \rho(\mathbf{r}, t) = \mu - V_{\text{ext}}(\mathbf{r}, t)$, which is identical to the Euler-Lagrange equation of equilibrium DFT, where the chemical potential, μ , keeps the particle number constant. In this limit the theory mimics “ultrafast” dynamics, where the density field instantaneously follows changes in the external potential.

(ii) *Ideal gas.* For a system of noninteracting particles the Helmholtz free energy is known exactly, $F_{\text{id}}[\rho] = k_B T \int d\mathbf{r} \rho(\mathbf{r}, t) [\ln(\rho(\mathbf{r}, t) \Lambda^3) - 1]$, where Λ is the thermal wavelength, and the dissipation functional is given by

$$P_{\text{id}}[\rho, \mathbf{J}] = \int d\mathbf{r} \frac{\gamma \mathbf{J}(\mathbf{r}, t)^2}{2\rho(\mathbf{r}, t)}. \quad (16)$$

Functional differentiation of this expression with respect to the current generates the (mean) friction force, such that $\mathbf{J}(\mathbf{r}, t) \cdot \delta P[\rho, \mathbf{J}] / \delta \mathbf{J}(\mathbf{r}, t) = \mathbf{J}(\mathbf{r}, t) \cdot \gamma \mathbf{v}(\mathbf{r}, t)$ represents

the dissipated power density resulting from the average motion of the system; here $\mathbf{v}(\mathbf{r}, t) = \mathbf{J}(\mathbf{r}, t)/\rho(\mathbf{r}, t)$ is the average velocity. The exact expression for the ideal current, $\gamma \mathbf{J}_{\text{id}}(\mathbf{r}, t)/\rho(\mathbf{r}, t) = -k_B T \nabla \ln \rho(\mathbf{r}, t) - \nabla V_{\text{ext}}(\mathbf{r}, t) + \mathbf{X}(\mathbf{r}, t)$ follows from substitution of (16) into (15).

(iii) *Dynamical density functional theory.* The intrinsic free energy functional of an interacting system can be written as $F[\rho] = F_{\text{id}}[\rho] + F_{\text{exc}}[\rho]$, where the excess contribution accounts for interparticle interactions. If we retain the full free energy, but assume that the dissipation is given by (16), then (15) yields $\mathbf{J}(\mathbf{r}, t) = \mathbf{J}_{\text{DDFT}}(\mathbf{r}, t)$, where

$$\frac{\gamma \mathbf{J}_{\text{DDFT}}(\mathbf{r}, t)}{\rho(\mathbf{r}, t)} = -\nabla \frac{\delta F[\rho]}{\delta \rho(\mathbf{r}, t)} - \nabla V_{\text{ext}}(\mathbf{r}, t) + \mathbf{X}(\mathbf{r}, t) \quad (17)$$

is precisely the current of DDFT [3, 4]. We thus gain new insight into the standard theory, namely that the adiabatic approximation is equivalent to assuming a trivial, noninteracting form for the dissipation functional. This observation suggests that superior theories can be obtained by developing approximations to $P[\rho, \mathbf{J}]$ which recognize the existence of interparticle interactions.

We can now go beyond DDFT by decomposing the dissipation power functional into two contributions,

$$P[\rho, \mathbf{J}] = P_{\text{id}}[\rho, \mathbf{J}] + P_{\text{exc}}[\rho, \mathbf{J}], \quad (18)$$

where $P_{\text{id}}[\rho, \mathbf{J}]$ is given by (16) and $P_{\text{exc}}[\rho, \mathbf{J}]$ accounts for dissipation arising from interparticle interactions. Microscopically, the excess dissipation is caused by correlated fluctuations of the particle velocities $\mathbf{v}_i(t)$ around the mean velocity $\mathbf{v}(\mathbf{r}_i(t), t)$. Using (18) the fundamental equation of motion (15) becomes

$$\begin{aligned} \frac{\gamma \mathbf{J}(\mathbf{r}, t)}{\rho(\mathbf{r}, t)} + \int dt' \frac{\delta P_{\text{exc}}[\rho', \mathbf{J}']}{\delta \mathbf{J}(\mathbf{r}, t)} = \\ -\nabla \frac{\delta F[\rho]}{\delta \rho(\mathbf{r}, t)} - \nabla V_{\text{ext}}(\mathbf{r}, t) + \mathbf{X}(\mathbf{r}, t). \end{aligned} \quad (19)$$

Corrections to DDFT thus come from the second term on the left hand side. It is instructive to compare (19) with the formally exact result obtained by integrating the many-body Smoluchowski equation over $N - 1$ particle coordinates [6]. Restricting ourselves for simplicity to systems that interact via pair potentials, $U = \sum_{i \neq j} \phi(\mathbf{r}_i, \mathbf{r}_j)$, an exact equation of motion is obtained,

$$\begin{aligned} \frac{\gamma \mathbf{J}(\mathbf{r}, t)}{\rho(\mathbf{r}, t)} = -k_B T \nabla \ln(\rho(\mathbf{r}, t) \Lambda^3) - \nabla V_{\text{ext}}(\mathbf{r}, t) \\ + \mathbf{X}(\mathbf{r}, t) - \int d\mathbf{r}' \frac{\rho^{(2)}(\mathbf{r}, \mathbf{r}', t)}{\rho(\mathbf{r}, t)} \nabla \phi(\mathbf{r}, \mathbf{r}'), \end{aligned} \quad (20)$$

where $\rho^{(2)}(\mathbf{r}, \mathbf{r}', t)$ is the exact (but unknown) out-of-equilibrium equal-time two-body density. Comparison

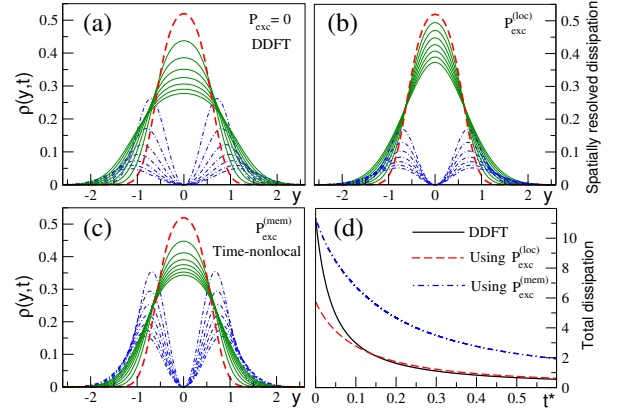


FIG. 1. Free relaxation of a density peak (slab geometry) from an initial state (red dashed curve) for which the density is confined to a parabolic potential. The solid green curves show the density and the blue dash-dotted curves show the spatially resolved dissipation (scaled down by a factor 20) for three different approximations to the excess dissipation (at dimensionless times $t^* \equiv tk_B T / \gamma d^2 = 0.2$ to 1.4 in steps of 0.2). a) Using DDFT, b) using (24), c) using (26). Panel d) shows how the total dissipated power decays as a function of time.

with (19) yields the identification

$$\begin{aligned} \int dt' \frac{\delta P_{\text{exc}}[\rho', \mathbf{J}']}{\delta \mathbf{J}(\mathbf{r}, t)} = \\ -\nabla \frac{\delta F_{\text{exc}}[\rho]}{\delta \rho(\mathbf{r}, t)} + \int d\mathbf{r}' \frac{\rho^{(2)}(\mathbf{r}, \mathbf{r}', t)}{\rho(\mathbf{r}, t)} \nabla \phi(\mathbf{r}, \mathbf{r}'). \end{aligned} \quad (21)$$

If we split the full nonequilibrium two-body density into an instantaneous-equilibrium and an irreducible part, $\rho^{(2)}(\mathbf{r}, \mathbf{r}', t) = \rho_{\text{eq}}^{(2)}(\mathbf{r}, \mathbf{r}', t) + \rho_{\text{irr}}^{(2)}(\mathbf{r}, \mathbf{r}', t)$, then we can identify the following two identities

$$\nabla \frac{\delta F_{\text{exc}}[\rho]}{\delta \rho(\mathbf{r}, t)} = \int d\mathbf{r}' \frac{\rho_{\text{eq}}^{(2)}(\mathbf{r}, \mathbf{r}', t)}{\rho(\mathbf{r}, t)} \nabla \phi(\mathbf{r}, \mathbf{r}'), \quad (22)$$

$$\int dt' \frac{\delta P_{\text{exc}}[\rho', \mathbf{J}']}{\delta \mathbf{J}(\mathbf{r}, t)} = \int d\mathbf{r}' \frac{\rho_{\text{irr}}^{(2)}(\mathbf{r}, \mathbf{r}', t)}{\rho(\mathbf{r}, t)} \nabla \phi(\mathbf{r}, \mathbf{r}'). \quad (23)$$

Equation (22) is an exact equilibrium relation [6], whereas (23) expresses the fact that the excess dissipation is intimately connected with beyond-adiabatic, irreducible two-point correlations. This is fully consistent with the interpretation that the dynamic functional is a generator for many-body induced friction forces in the system.

Any practical application of the framework, i.e. implementation of (19) along with (3), requires one to choose an approximation for $P_{\text{exc}}[\rho, \mathbf{J}]$ for the given physical system (as characterized by U). Here we discuss several model approximations to P_{exc} , the simplest of which is given by the space- and time-local expression

$$P_{\text{exc}}^{(\text{loc})}[\rho, \mathbf{J}] = \int d\mathbf{r} (\tilde{\gamma}(\rho) - \gamma) \frac{\mathbf{J}(\mathbf{r}, t)^2}{2\rho(\mathbf{r}, t)}, \quad (24)$$

where $\tilde{\gamma}(\rho)$ is a density-dependent friction factor which can be approximated using either a virial expansion or, in the case of hard spheres, by employing closed form expressions [12]. Using (24) slows the dynamics in regions of high density relative to DDFT [13], but will still fail to describe collision induced dissipation arising from shearing motions in the fluid. To capture this requires a nonlocal treatment in space and in time which recognizes the finite range of the colloidal interactions. A suitably general form is given by

$$P_{\text{exc}}^{(\text{nloc})}[\rho, \mathbf{J}] = \int d\mathbf{r} \int d\mathbf{r}' \int_{t_0}^t dt' \mathbf{J}(\mathbf{r}, t) \cdot \mathbf{K}(\mathbf{r}, t; \mathbf{r}', t') \cdot \mathbf{J}(\mathbf{r}', t'), \quad (25)$$

where the convolution kernel \mathbf{K} is a second-rank tensor describing current-current scattering and is in general a functional of ρ . The possibility of incorporating temporal nonlocality represents a powerful feature of the present approach. A simple specific form for the scattering kernel which captures this is $\mathbf{K} = \gamma m(t-t')\delta(\mathbf{r}-\mathbf{r}')/\rho(\mathbf{r}, t)$, where $m(t-t')$ is a memory function (with units of inverse time). As a result we obtain a spatially local excess dissipation functional with temporal memory, given by

$$P_{\text{exc}}^{(\text{mem})}[\rho, \mathbf{J}] = \int d\mathbf{r} \int_{t_0}^t dt' \frac{\gamma \mathbf{J}(\mathbf{r}, t) \cdot \mathbf{J}(\mathbf{r}, t')}{\rho(\mathbf{r}, t)} m(t-t'). \quad (26)$$

For general interactions and time-dependent external fields the memory need not be time translationally invariant, as assumed here. The equation of motion (19) thus becomes

$$\frac{\gamma}{\rho(\mathbf{r}, t)} \left(\mathbf{J}(\mathbf{r}, t) + \int_{t_0}^t dt' \mathbf{J}(\mathbf{r}, t') m(t-t') \right) = -\nabla \frac{\delta F[\rho]}{\delta \rho(\mathbf{r}, t)} - \nabla V_{\text{ext}}(\mathbf{r}, t) + \mathbf{X}(\mathbf{r}, t). \quad (27)$$

We have performed numerical calculations using the (three-dimensional) hard sphere system in a slab geometry using approximations (24) and (26). The (spatially non-local) Rosenfeld functional [14] was used to approximate $F_{\text{exc}}[\rho]$. The system is initially confined along the y -axis by a parabolic potential, $V_{\text{ext}}(\mathbf{r}, t < 0) = by^2/2$, where b is a constant measuring the strength of the trapping potential. We then switch off the confinement at $t = t_0 = 0$, such that $V_{\text{ext}}(\mathbf{r}, t > 0) = 0$, and monitor the time evolution of the density. In Fig. 1 we show the density calculated using three approximations: DDFT, the density dependent friction coefficient (24) (where we have employed the expression for $\tilde{\gamma}(\rho)$ from [12]) and, finally, the temporally nonlocal approximation (26). When implementing (26) we have approximated the memory function by the simple form $m(t-t') = (\alpha/\tau) \exp(-(t-t')/\tau)$, where α is a dimensionless parameter and τ is a relaxation time.

The density profiles in Fig. 1 (generated using the parameter values $\alpha = 0.8, b = 10k_B T, \tau = 0.05\gamma d^2/k_B T$, where the particle diameter, d , sets the length-scale) show that both (24) and (26) slow the dynamics. The magnitude of the retardation achieved using (26) depends upon the choice of parameters α and τ determining the memory. While (26) generates density profiles with a similar functional form to those from DDFT, Eq. (24) leads to a density more sharply peaked at the origin. The spatially resolved dissipation (blue dash-dotted curves) confirms the intuition that power is dissipated mostly in regions of high density gradient and the spatial integral of this quantity, $\int d\mathbf{r} \mathbf{J}(\mathbf{r}, t) \cdot \delta P[\rho, \mathbf{J}]/\delta \mathbf{J}(\mathbf{r}, t)$, decays towards zero as the system approaches equilibrium. It is well known that relaxation rates predicted by standard DDFT are significantly faster than those found in simulation [4, 13]. On the basis of our findings we conclude that this failing can be remedied by the incorporation of temporal nonlocality in the excess dissipation functional, leading to memory effects in the equation of motion for $\mathbf{J}(\mathbf{r}, t)$. If an exponential memory is employed then the computational demands are comparable to those of DDFT.

In summary, we have shown that collective Brownian dynamics can be formulated as a variational theory (14) based on the dissipative power as a functional of the one-body density and the one-body current. The generalization to mixtures of different species is straightforward: the ideal contribution becomes $\sum_i P_{\text{id}}[\rho_i, \mathbf{J}_i]$, where i enumerates the different species, whereas $P_{\text{exc}}[\{\rho_i\}, \{\mathbf{J}_i\}]$ generates dynamical coupling between particles of different species [15].

The appeal of our approach stems from its utility and ease with which it can be implemented, which contrast strongly with the time-dependent DFT formalism by Chan and Finken [16]. As far as we are aware, the approach of [16] has never been applied to any model system, possibly because it is built around an action functional, which could not (yet) been approximated in any systematic or physically intuitive way. This is not the case with the excess dissipation functional identified in the present work, for which even simple expressions, such as (26), transcend the adiabatic approximation. While DDFT remains an active field of research (e.g. hydrodynamic interactions [17] and arbitrary particle shapes [18] have very recently been addressed), it is important to appreciate that all extensions and modifications proposed since the original presentation of DDFT [3] have been firmly under the constraint of adiabaticity. The power functional approach is free of this restriction.

Finally, the relationships (8) and (9) of the one-body fields to their generating functional imply that two- and higher-body dynamic correlation functions can be generated from further functional differentiation, putting a nonequilibrium Ornstein-Zernike relation within reach. Work along these lines, as well as applications to driven

lattice models [19], is currently in progress.

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